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Influence of structural transition in metal on charge transport in nanocrystal metal-polymer-metal system

A. N. Lachinov†, T. G. Zagurenko†, V. M. Kornilov† and R. Z. Valiev‡

† Institute of Physics of Molecules and Crystals,
 Ufa Research Center, RAS, 450000 Ufa, Russia.

‡ Institute of Physics of Advanced Materials, UGATU,
 450000, Ufa, Russia.

The paper presents the results of the investigation of influence of the structural transitions in nanocrystal metals on the electrical conductivity in a nanocrystal metal-polymer-metal system (NM-P-M). The experiment idea was as follows. The structural transitions in the nanocrystal metal induced by a magnification of its grain size at specific temperatures should lead to a modification (decreasing) of the work function ϕ_m . When such a metal is in contact with the electroactive polymer, the change in ϕ_m should lead to a change in the polymer space charge and as a result to a change in the charge carrier injection conditions. In the present work the nanocrystal metals (Cu, Ni) were selected as electrodes. The nanocrystal state is nonequilibrium in these metals and the rather small heating to $T \approx 0.4T_{cr}$ gives rise to restoring of the initial structure [1]. The nanocrystal structure was identified by methods of the electronic microscopy and X-ray structure analysis. As it was shown earlier [2], an ultra fine-grained structure is formed in Cu and Ni as a result of the intensive plastic deformation of torsion (IPD). The resulting nanocrystal structure has a size of grains of about 100 nm, high level of the crystal lattice microdistortions, nonequilibrium grain boundaries, high density of the grain-boundary dislocations, increased static and dynamic atomic displacements. During the last years the IPD method was successfully used for obtaining the nanostructures in various metals and alloys [1, 2]. IPD can cause the modification of the material work function on 0.1 eV and more [1, 3]. It was expected, that the heating would induce restoring the initial ϕ_m value, which will affect the electrical current in NM-P-M system. The measurements of the current were carried out using the technique described in [4]. As a polymer the films of polyphthalidilidenbiphenylilene (PPB) of about 0.8 μm thick were used. This polymer reveals insulator-metal transition.

Figure 1 shows the temperature dependence of the current $I(T)$ flowing through a NM-P-M system. When the copper is used as NM, the raise in temperature does not change the current up to $T_1 = 145^\circ\text{C}$. In the range of temperatures $T_1 = 145^\circ\text{C} - T_2 = 175^\circ\text{C}$ $I(T)$ dependence reveals a peak with a maximum at $T_{\text{max}} \approx 160^\circ\text{C}$. The similar behavior is observed when the nanocrystal Ni is used as the electrode. In the latter case the maximum of the current is observed at $T_{\text{max}} \approx 215^\circ\text{C}$. The similar experiments were carried out using Cu and Ni with equilibrium grain structure instead of nonequilibrium nanocrystal samples. In these experiments the following samples were used:

1. The initial large-grain metal, from which afterwards the nanocrystal samples were obtained.
2. Heat-treated nanocrystal samples. (The samples were treated during 30 min at 500°C , which, as is known [1], completely destroys the nanocrystal structure.) The measurements revealed no temperature singularities in the system conductivity in the investigated tem-

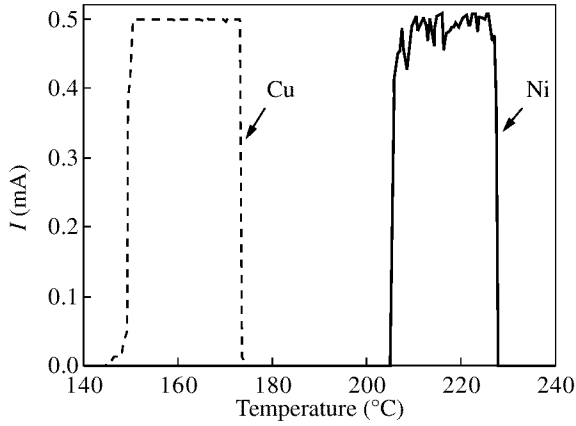


Fig. 1. Temperature dependence of current flowing through the polymer film. Solid line—upper electrode is nanostructural Cu, dashed line—nanostructural Ni. Polymer film thickness 2 μm ; lower electrode V; applied voltage 5 V; heating rate 8°C/min.

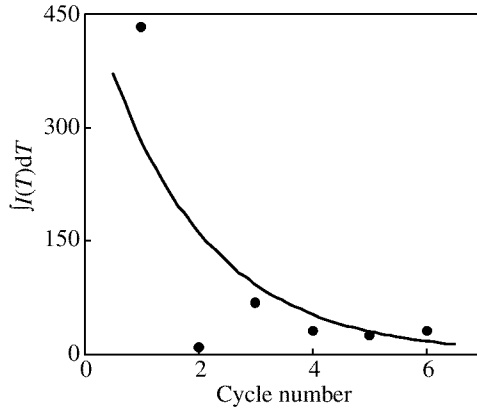


Fig. 2. Integral $I(T)$ —cycle number dependence. Film thickness 2 μm ; upper electrode—nanostructural Cu; lower electrode V; applied voltage 5 V; heating rate 8°C/min.

perature range. The microstructure evolution of the nanocrystal Cu and Ni during heating was explicitly investigated in papers [3, 5] using mentioned above methods and also using differential scanning calorimetry technique. The results obtained testify to intensive relaxation processes associated with a transformation of the defect structure during low temperature heat-treatment. These processes are observed in the temperature ranges of 100–200°C for Cu and of 180–220°C for Ni. These temperature ranges are close to those, in which mentioned above conductivity peculiarities are detected. It allows us to make a conclusion that the mechanisms initiating these modifications are similar and are caused by the temperature transformation of the microstructure of the metal samples.

Multiple measurements of $I(T)$ dependence on the same sample showed that with the increase in the measurement cycles the decrease in the amount of the charge flowing through the sample is observed down to some minimal value which corresponds to the

current flowing through the sample at room temperature. It, apparently, reflects that fact, that for identical conditions of measurements in multiple cycles, the relative changes in the work function decrease (1,2) as the sample structure approaches equilibrium state. It is necessary to pay attention to some peculiarities of the dependence represented in Fig. 2.

1. During one cycle of heating the sample does not return to the equilibrium state. The full transition occurs during several cycles.
2. As a rule, the maximum characteristic temperature, at which the current singularity is registered, is observed in the first cycles of measurements.
3. With the increase in the number of cycles the downward tendency for the characteristic temperature is observed. Simultaneously the extension of the temperature range occurs, in which the effect is observed. In accordance with (1, 2) in addition to the work function the electrical field should influence the injection current. This influence was estimated by integrating $IU(T)$ on dT in the range of (T_0, T_n) depending on the voltage applied. This integral is proportional to the net charge, passing through the sample in the given temperature range. T_0 and T_n —boundary temperatures of the experimental interval, $IU(T)$ —the temperature dependence of the current at voltage U on the electrodes. The analysis of the experimental results showed, that for $U < 0.1$ V the magnitude of the charge flowing through the sample is constant. In the range of voltages of $0.1 \text{ V} < U < 5 \text{ V}$ superlinear increase in the net charge passing through the polymer is observed. Above 5 V the dependence approaches the linear. The similar dependence is observed practically on all samples. The investigations performed indicate, that the use of a thin polymer film of PPB type in M-P-M heterostructure allows to study surface transformations in metal electrodes caused by structural transitions. This method is very sensitive, simple in a realization and consequently can appear rather effective.

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